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(54) **Directly paintable thermoplastic polyolefin composition containing oxidized polyethylene waxes**

Lackierbare thermoplastische Elastomerezusammensetzung, welche oxidierte Polyethylenwachse enthält

Composition élastomère thermoplastique pouvant être peinte contenant des cires oxydées à base de polyéthylène

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(56) References cited:  
**EP-A- 0 292 264 EP-A- 0 389 135**  
**WO-A-98/02490 DD-A- 214 851**

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**Description**Field of the Invention

- 5 [0001] This invention relates to thermoplastic polyolefin compositions containing polymers grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid, and functionalized polymeric additives.

Background of the Invention

- 10 [0002] Thermoplastic polyolefins (TPOs) are uncrosslinked blends of olefin polymers and polyolefin elastomers. They can be made by physically blending in an internal mixer, or by polymerizing in a reactor. These materials are not paintable or coatable, because the paints or coatings consist of polar materials like urethanes, acrylics, epoxies, or melamines that have very poor adhesion to nonpolar materials like polyolefins. Typically an adhesion promoter is used as the tie layer between the TPO substrate and the paint coating. This extra step adds to the cost of the product, and the coating is not very durable.

- 15 [0003] European Patent Application 662496 discloses a paintable or printable polymer composition consisting of a polyolefin or polyolefin/rubber blend and 0.1 to 10% by weight of at least one polymeric additive that is the reaction product of (a) a polyolefin or polyester modified by an unsaturated acid, ester or anhydride, and (b) an amine-, hydroxy-, or alkoxy-terminated polyoxyethylene, polyoxypropylene, or a copolymer of the two, e.g., the reaction product of a maleic anhydride-modified polypropylene wax and a methoxy-capped poly(ethylene oxide) glycol.

- 20 [0004] European Patent Application 634424 discloses a blend of polypropylene with the reaction product of maleated polypropylene and a polyether amine. The blend displays improved paintability, improved impact resistance, and excellent mold flowability compared to blends of polypropylene and the reaction product of polypropylene and maleated polypropylene. However, the compositions disclosed in EP 662496 and EP 634424 are both lacking in durability. Blends having similar composition and properties are disclosed also in the International Patent Application WO98/02490.

- 25 [0005] Thus, there is still a need for a TPO composition that can meet the more stringent requirements for paint adhesion and durability that are required in today's marketplace, particularly in the automotive industry.

Summary of the Invention

- 30 [0006] The composition of this invention comprises, by weight:

- (1) 100 parts of a thermoplastic polyolefin comprising an olefin polymer having an isotactic index of at least 80 and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%;
- 35 (2) 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a propylene homopolymer or propylene copolymer with ethylene or a  $C_4$ - $C_8$   $\alpha$ -olefin having an ethylene or  $\alpha$ -olefin content of 0.5% to 20%, grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid and having an anhydride content of 2% to 5%;
- (3) 3 to 20 parts per hundred parts of the thermoplastic polyolefin of an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40;
- 40 (4) a functionalized polymer that is reactive with the anhydride groups of the grafted polymers, selected from the group consisting of:

- (a) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated polyalkylene glycol;
- (b) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyolefin;
- 45 (c) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polybutadiene;
- (d) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated olefin/alkylene oxide copolymer;
- (e) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyalkylene oxide;
- (f) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a methoxy-terminated polyalkylene oxide;
- 50 (g) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated olefin/alkylene oxide copolymer; and
- (h) mixtures thereof;

- (5) optionally, 5 to 30 parts per hundred parts of the thermoplastic polyolefin of a polyolefin rubber grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid, having an anhydride content of at least 0.3% but less than 3% and comprising a polymer of ethylene and a  $C_3$ - $C_8$   $\alpha$ -olefin, optionally containing 0.5% to 10% of a diene, having an ethylene content of 30% to 70%; and

- 55 (6) optionally, 5 to 20 parts per hundred parts of the thermoplastic polyolefin of an ethylene polymer grafted with

an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid, having an anhydride content of 1% to 16% and a molecular weight  $M_n$  of 500 to 5000, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic elastomer are also present.

[0007] Injection molded parts such as automobile bumpers made from this composition are directly paintable with polar paints or coatings without the need for a layer of adhesion promoter between the thermoplastic polyolefin surface and the paint, and exhibit excellent paint adhesion and durability.

#### Detailed Description of the Invention

[0008] Component (1) of the composition of this invention is a thermoplastic polyolefin comprising a crystalline olefin polymer and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%. Suitable thermoplastic polyolefins include, for example,

(a) a composition comprising, by weight,

(i) 10% to 60%, preferably 20% to 50%, of a propylene homopolymer having an isotactic index greater than 90, preferably between 95 and 98, or a crystalline propylene copolymer with ethylene and/or a  $C_{4-8}$   $\alpha$ -olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(ii) 30% to 60%, preferably 30% to 50%, of an amorphous ethylene-propylene or ethylene-butene copolymer, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and has an ethylene content of 30% to 70%;

(iii) 2% to 20%, preferably 7% to 15%, of a semi-crystalline ethylene-propylene or ethylene-butene copolymer that is xylene insoluble at 25°C and has an ethylene content of greater than 75% but less than 92%; and

(iv) 5% to 20%, preferably 7% to 15%, of an ethylene polymer having a density of 0.91 to 0.96 g/cm<sup>3</sup> and a melt index of 0.1 to 100 g/10 min, preferably 15 to 50 g/10 min. Ethylene homopolymer is preferred. However, copolymers containing 8% or less of an  $\alpha$ -olefin comonomer can also be used.

(b) a composition comprising, by weight:

(i) 20% to 70%, preferably 50% to 70%, of a crystalline propylene homopolymer having an isotactic index greater than 90, preferably between 95 and 98, or a crystalline propylene copolymer with ethylene and/or a  $C_4-C_8$   $\alpha$ -olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(ii) 20% to 75%, preferably 30% to 50%, most preferably 30% to 35%, of an amorphous copolymer of ethylene selected from the group consisting of (1) ethylene/propylene, (2) ethylene/butene-1, (3) ethylene/octene-1, and (4) mixtures thereof, optionally containing 1% to 10%, preferably 1% to 4%, of a diene, which is xylene soluble at 25°C and has an ethylene content of 30% to 70%, preferably 40% to 60%; and

(iii) 2% to 30%, preferably 2% to 10%, most preferably 2% to 5%, of a semi-crystalline copolymer of ethylene selected from the group consisting of (1) ethylene/propylene, (2) ethylene/butene-1, (3) ethylene/octene-1, and (4) mixtures thereof, which is xylene insoluble at room temperature and has an ethylene content of greater than 90%;

(c) a composition comprising, by weight,

(i) at least one heterophasic polyolefin composition comprising:

(1) 90% to 55% of a propylene polymer material selected from the group consisting of a propylene homopolymer having an isotactic index greater than 90, and a crystalline copolymer of propylene and an  $\alpha$ -olefin of the formula  $CH_2=CHR$ , where R is H or  $C_2-C_8$  alkyl, the  $\alpha$ -olefin being less than 10% of the copolymer, and

(2) 10% to 45% of an elastomeric copolymer of propylene and an  $\alpha$ -olefin of the formula  $CH_2=CHR$ , where R is H or  $C_2-C_8$  alkyl, the  $\alpha$ -olefin being 50% to 70% of the elastomeric copolymer, and 10% to 40% of the elastomeric copolymer being insoluble in xylene at ambient temperature, and

(ii) 5 to 50 parts, preferably 10 to 30 parts, and most preferably 10 to 25 parts, per hundred parts of (c)(i) of an elastomeric copolymer of ethylene and a  $C_3-C_8$   $\alpha$ -olefin made with a metallocene catalyst. If more than one heterophasic polyolefin (c)(i) is present, the heterophasic polyolefins can be combined in any proportion.

(d) a composition comprising, by weight:

(i) 30% to 50%, preferably 35% to 45%, of a propylene homopolymer having an isotactic index greater than 90, and

(ii) 70% to 50%, preferably 65% to 55%, of an olefin polymer composition comprising:

(1) 25% to 50% of a crystalline propylene homopolymer with a solubility in xylene at room temperature of less than or equal to 4%, or a crystalline copolymer of propylene with ethylene or a C<sub>4</sub>-C<sub>8</sub> α-olefin having an ethylene or α-olefin content of 0.5% to 3%, and a solubility in xylene at room temperature of less than or equal to 4%, and

(2) 50% to 75% of an amorphous copolymer of ethylene and a C<sub>4</sub>-C<sub>8</sub> α-olefin, wherein the α-olefin content is 10% to 20%, and the copolymer is 10% to 40% soluble in xylene at room temperature; and

(e) a composition comprising, by weight:

(i) 80% to 30%, preferably 70% to 50%, of a propylene homopolymer having an isotactic index greater than 90, and

(ii) 20% to 70%, preferably 30% to 50%, of an elastomeric copolymer of ethylene and a C<sub>3</sub>-C<sub>8</sub> α-olefin, optionally containing 1% to 10%, preferably 1% to 4%, of a diene, and having an ethylene content of 30% to 70%, preferably 40% to 60%.

Thermoplastic polyolefins (a) and (c) are preferred.

[0009] Thermoplastic polyolefins (a) and (b) and compositions (c)(i) and (d)(ii) are typically prepared by sequential polymerization in at least two stages. Alternatively, the components can be prepared separately and then blended together by melt-kneading or melt blending. The polymerization conditions and the polymerization catalyst are described in more detail in U.S. Patents 5,143,978, 5,302,454, 5,360,868 and 5,486,419. Sequential polymerization is preferred.

[0010] For TPO (a), (i) can be made in the first reactor, (ii) and (iii) in the second reactor, and (iv) in the third reactor. Alternatively, (iv) can be made in the second reactor and (ii) and (iii) in the third reactor.

[0011] For TPO (b), (i), (ii), and (iii) are preferably formed in a reactor or series of reactors in at least two stages by first polymerizing propylene to form (i) and then polymerizing ethylene and propylene, butene-1, or octene-1, or mixtures thereof, in the presence of (i) and the catalyst used in the first stage to form (ii) and (iii). The polymerization can be conducted in the liquid or gas phase or in liquid-gas phase.

[0012] For TPO (b), (i) can be prepared using a Ziegler-Natta catalyst or a mixture of Ziegler-Natta and metallocene catalysts. Components (ii) and (iii) can be prepared using Ziegler-Natta or metallocene catalysts or a combination of the two, with one type of catalyst being used for one stage and the other type of catalyst being used for the next stage when the TPO is made by sequential polymerization.

[0013] The C<sub>4</sub>-C<sub>8</sub> α-olefins useful in the preparation of the thermoplastic polyolefins include, for example, butene-1, pentene-1, hexene-1, 4-methylpentene-1 and octene-1.

[0014] The diene, when present, is typically 1,3-butadiene, 1,4-hexadiene, 1,5-hexadiene or ethylidenenorbornene.

[0015] Component (2) is a propylene homopolymer or propylene copolymer with ethylene or a C<sub>4</sub>-C<sub>8</sub> α-olefin, grafted with an anhydride of an aliphatic α,β-unsaturated dicarboxylic acid and having an ethylene or α-olefin content of 0.5% to 20%, preferably 1% to 10%, and most preferably 1% to 5%. The polymer has an anhydride content of 2% to 5%, preferably 3% to 4%, and preferably has a molecular weight M<sub>n</sub> of 2500 to 25,000, more preferably 3000 to 10,000. Maleic anhydride is the preferred anhydride. Component (2) is present in an amount of 5 to 20 parts, preferably 8 to 16 parts, most preferably 10 to 14 parts, per hundred parts of the thermoplastic polyolefin.

[0016] Component (3) is an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40. The oxidized wax is present in an amount of 3 to 20 parts, preferably 5 to 15 parts, most preferably 5 to 10 parts, per hundred parts of the thermoplastic polyolefin, whether or not the maleic anhydride-grafted rubber is present.

[0017] Component (4) is a functionalized polymer that is reactive with the anhydride groups of the grafted polymers (2) and, when present, (5) and (6), selected from the group consisting of (a) an amine-terminated polyalkylene glycol, (b) a hydroxy-terminated polyolefin, (c) a hydroxy-terminated polybutadiene, (d) hydroxy-terminated olefin/alkylene oxide copolymers, (e) hydroxy-terminated polyalkylene oxides, (f) methoxy-terminated polyalkylene oxides, (g) amine-terminated olefin/alkylene oxide copolymers, and (h) mixtures thereof.

[0018] Component (4)(a), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin. The polyalkylene glycol can be, for example, polyethylene glycol, polypropylene glycol, copolymers of ethylene glycol and propylene glycol, poly(1,2-butylene glycol) and poly(tetramethylene glycol).

[0019] Component (4)(b), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin. Polyethylene is the preferred polyolefin, although polypropylene, polybutene, and copolymers of ethylene and another  $\alpha$ -olefin can also be used.

[0020] Component (4)(c), when present, is used in an amount of 2 to 6 parts, preferably 2 to 4 parts, per hundred parts of the thermoplastic polyolefin.

[0021] Component (4)(d), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. An ethylene/ethylene oxide copolymer is preferred, although other copolymers such as ethylene/propylene oxide, propylene/ethylene oxide, butene/ethylene oxide and butene/propylene oxide copolymers can also be used. The amount of alkylene oxide can be from 10% to 99.9%, preferably 50% to 98%, and most preferably 75% to 95%, based on 100% of the copolymer.

[0022] Component (4)(e), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. Polyethylene glycol is preferred, however polypropylene glycol, copolymers of ethylene glycol and propylene glycol, poly(1,2-butylene glycol) and poly(tetramethylene glycol) can also be used.

[0023] Component (4)(f), when present, is used in an amount of 2 to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. Suitable polyalkylene oxides are those described under (4)(e).

[0024] Component (4)(g), when present, is used in an amount of 2 parts to 8 parts, preferably 2 to 6 parts, per hundred parts of the thermoplastic polyolefin. The amount of alkylene oxide can be 10% to 99.9%, preferably 50% to 98%, and most preferably 75% to 95%. Examples of suitable olefin/alkylene oxide copolymers are described under (4)(d).

[0025] When using a combination of functionalized polymers, the amount of each component can vary widely from 0.1 % to 99.9% of each, based on the total amount of functionalized polymers. It is preferred that one component be present in an amount higher than 50%, preferably higher than 60%, based on the total amount of functionalized polymers.

[0026] Instead of adding the functionalized polymer directly to the thermoplastic polyolefin, an adduct of the functionalized polymer and the anhydride-grafted polypropylene or ethylene/propylene copolymer can be prepared separately, then blended with the thermoplastic polyolefin.

[0027] Optional component (5) is a polyolefin rubber grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid and comprising a polymer of ethylene and a  $C_3$ - $C_8$   $\alpha$ -olefin, optionally containing 0.5% to 10% of a diene, preferably 2% to 6%. The anhydride-grafted polyolefin rubber has an ethylene content of 30% to 70%, preferably 40% to 60%, and has an anhydride content of at least 0.3% but less than 3%. Maleic anhydride is the preferred anhydride. When present, the anhydride-grafted rubber is used in an amount of 5 to 30 parts, preferably 5 to 15 parts, most preferably 5 to 12 parts, per hundred parts of the thermoplastic polyolefin.

[0028] Optional component (6) is an ethylene polymer grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid and having an anhydride content of 1% to 16% by weight, preferably 2% to 13%, most preferably 3% to 13%. Maleic anhydride is the preferred anhydride. Ethylene homopolymer is preferred. However, copolymers containing 10% or less of an  $\alpha$ -olefin comonomer can also be used. The ethylene polymer preferably has a molecular weight  $M_n$  of 500 to 5000, preferably 600 to 2000, most preferably 600 to 1000. When component (6) is used, it is present in an amount of 5 to 20 parts, preferably 5 to 10 parts, per hundred parts of the thermoplastic polyolefin, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic polyolefin are also present.

[0029] The composition of the present invention can also contain other conventional additives, for example, antioxidants; stabilizers; extender oils such as paraffinic and naphthenic oils; fillers such as  $CaCO_3$ , talc, carbon black and zinc oxide, or flame retardants.

[0030] If non-polymeric additives such as conductive or non-conductive carbon black are used, they are preferably added after the functionalized polymer has reacted with the anhydride-grafted polymers. The additives can also be added as a dispersion in a polymer, preferably an olefin polymer.

[0031] The compounding or melt blending of the components of the composition can be carried out on an open roll, in an internal mixer (Banbury or Haake mixers), or in single-screw or twin screw extruders.

[0032] The compositions of this invention can be formed in any way, such as, for example, by extrusion, compression molding and thermoforming. Injection molding is preferred. They can also be co-extruded or co-injection molded with other polyolefin materials such as propylene homopolymers, copolymers and graft copolymers; ethylene homopolymers and copolymers, or thermoplastic polyolefins such as those described previously. They can also be co-extruded or co-injection molded with olefin-based dynamically vulcanized elastomers or olefin-compatible thermoplastic elastomers such as styrene/butadiene copolymers.

[0033] In order to create a more severe test for paint adhesion, the specimens for testing were prepared using a pin-gated mold rather than the fan-gated mold typically used for molding thermoplastic polyolefins. Durability depends upon the paint thickness - the thicker the paint or film, the better the durability. In the following examples and comparative examples only one coat of paint was used with an approximately 30.5  $\mu m$  (1.2 mil) film thickness, which is a very severe

test. A typical durability test used in the automotive industry also employs a top coat that has a low coefficient of friction, which reduces the severity of the test. No top coat was used in the following examples and comparative examples.

[0034] The samples for testing were prepared by dry blending the ingredients and reactive mixing in a twin screw extruder at a temperature of 232°C (450°F) and pelletizing the resultant material. The pellets were injection molded into disks that were painted with a 30.5 µm to 51 µm (1.2 to 2 mil) thick coating using DuPont 872 white paint and cured at 121°C (250°F) for thirty minutes. A lattice pattern of squares with each square 6.2 mm (¼ inch) in size was scribed on the painted disk at the end opposite the gate area of the disk. Adhesive tape (3M 898) was pressed onto the paint and pulled off to test the amount of paint removed or the paint adhesion. The % failure was recorded as the % of the squares removed by the tape after one pull. The durability was determined by using a Taber abrader with a type C scuffing head assembly and a one pound load. The painted disk was placed in an oven at 70°C for one hour, removed and placed on the platform of the abrader. The scuffing head was placed in contact with the painted surface and the disk was rotated for a specified number of cycles. The amount of paint removed from the complete circumference subtended by the scuffing head was recorded as the % failure.

[0035] The criteria set for acceptable paint adhesion were <50% failure in the gate area of the disk and <10% in the area opposite the gate area after the first pull, and <85% failure in the gate area and <50% in the area opposite the gate after the fifth pull. The criteria for satisfactory durability was <50% failure after 100 cycles.

[0036] In this specification all parts and percentages are by weight unless otherwise noted.

#### Examples 1-4 and Comparative Examples 1-3

[0037] Examples 1-4 show the paint adhesion and durability of compositions containing a thermoplastic polyolefin (TPO), a maleic anhydride-grafted polypropylene (MA-g-PP), an oxidized polyethylene wax, and an amine-terminated polyethylene oxide (ATPEO), with and without a maleic anhydride-grafted polyolefin rubber (MA-g-rubber). The effect of the melting point of the oxidized polyethylene wax was also demonstrated. The results are shown in Table 1.

[0038] In Table 1, TPO 1 contained 55% propylene homopolymer, 3% semi-crystalline ethylene/propylene copolymer that had a propylene content of ~10% and was insoluble in xylene at room temperature, 30% amorphous ethylene/propylene copolymer rubber that had an ethylene content of 50% and was soluble in xylene at room temperature, and 12% ethylene homopolymer having a melt index of 50 g/10 min.

[0039] MA-g-rubber 1 was Exxelor™ VA-1803 ethylene/propylene rubber containing 0.7% grafted maleic anhydride, commercially available from Exxon Chemical Company. MA-g-rubber 2 was Royaltuf 465A ethylene/propylene/non-conjugated diene terpolymer rubber containing 0.4% grafted maleic anhydride, having a Mooney viscosity (ML 1 + 4 at 125°C) of 60 and an ethylene/propylene ratio of 75/25, commercially available from Uniroyal Chemical Company, Inc.

[0040] MA-g-PP was Epolene™ E-43 maleic anhydride-modified polypropylene wax, commercially available from Eastman Chemical Company.

[0041] The ATPEO was XTJ-418 monoamine-terminated polyethylene oxide, commercially available from Huntsman Corporation.

[0042] The antioxidant was Irganox™ B 225, a blend of 1 part Irganox™ 1010 tetrakis[methylene(3,5-di-tert-butyl-4-hydroxyhydrocinnamate)methane and 1 part Irgafos™ 168 tris(2,4-di-tert-butylphenyl) phosphite antioxidant, commercially available from CIBA Specialty Chemicals Company.

[0043] Petrolite™ C-3500 oxidized polyethylene wax, Petrolite™ C-9500 oxidized polyethylene wax, and Petrolite™ E-2020 oxidized polyethylene wax are commercially available from Petrolite Corporation. The melting points and other properties of the waxes are given in Table 2. In Table 2, the numbers in parentheses are estimated values.

Table 1

	Ex. 1	Ex. 2	Ex. 3	Comp. 1	Comp. 2	Comp. 3
TPO 1	100	100	100	100	100	100
MA-g-rubber 1	5	5	--	5	--	5
MA-g-rubber 2	5	5	--	5	--	5
MA-g-PP	10	10	10	10	10	10
Petr. C-3500	5	--	10	--	--	--
Petr. C-9500	--	5	--	--	--	--
Pett. E-2020	--	--	--	--	10	5
ATPEO	3	3	3	3	3	3

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Table 1 (continued)

	Ex. 1	Ex. 2	Ex. 3	Comp. 1	Comp. 2	Comp. 3
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion						
%Failure	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	12/0	42/3	12/6	64/0	64/18	48/18
3 <sup>rd</sup> pull	30/8	48/9	36/12	80/40	76/42	56/48
5 <sup>th</sup> pull	46/36	70/25	50/36	94/65	94/66	84/68
Durability % Failure						
25 cycles	0	0	0	25	14	40
100 cycles	6	22	20	60	50	80

Table 2

	Acid No.	Mn	Viscosity at @ 149°C mPa.s	Density g/cm <sup>3</sup>	Melt Index g/10 min	Melt Pt. °C
AC 307	5-9	50,000	85,000	0.98	>1,000	140
AC 325	25	6,000	4,400	0.99	>2,000	136
Petr. C-8500	9	(8,000)	(6,000)	--	>2,000	95
Petr. C-7500	15	(6,000)	(4,000)	--	>3,000	97
Petr. C-3500	24	1,500	30	--	>5,000	96
Petr. C-9500	31	(1,000)	(20)	--	>5,000	94
Petr. E-2020	22	2,500	75	--	>5,000	116
Petr. E-1040	40	(1,500)	25	--	>5,000	106

[0044] Examples 1 and 2 show the improvement in paint adhesion and durability when an oxidized polyethylene wax is used along with the TPO, MA-g-PP, MA-g-rubber, and the ATPEO. Comparative Example 1 does not contain an oxidized polyethylene wax. Examples 3 and 4 show that the oxidized waxes produce good paint adhesion and durability without the MA-g-rubber. Comparative Examples 2 and 3 show that not all oxidized polyethylene waxes are effective in the compositions of this invention.

Examples 5-7 and Comparative Examples 4-6

[0045] These examples and comparative examples show the effect of using oxidized polyethylene waxes with varying melting points on paint adhesion and durability. All formulations contain MA-g-rubbers, MA-g-PP, and an ATPEO in addition to the TPO.

[0046] TPO 1, the MA-g-rubbers, the MA-g-PP, the antioxidant, and the ATPEO were the same as in Examples 1-4.

[0047] The results are shown in Table 3. The melting points and other properties of the waxes are given in Table 2.

Table 3

	Comp. 4	Comp. 5	Comp. 6	Ex. 5	Ex. 6	Ex. 7
TPO 1	100	100	100	100	100	100
MA-g-rubber 1	5	5	5	5	5	5
MA-g-rubber 2	5	5	5	5	5	5
MA-g-PP	10	10	10	10	10	10
Petr. E-1040	--	-	5	--	--	--

Table 3 (continued)

	Comp. 4	Comp. 5	Comp. 6	Ex. 5	Ex. 6	Ex. 7
Petr. C-3500	--	--	--	10	--	--
Petr. C-7500	--	--	--	--	5	--
Petr. C-8500	--	--	--	--	--	5
AC 307	5	--	--	--	--	--
AC 325	--	5	--	--	--	--
ATPEO	3	3	3	3	3	3
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion						
% Failure	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	80/80	90/99	18/12	6/0	0/0	36/0
3 <sup>rd</sup> pull			84/36	30/0	78/0	28/12
5 <sup>th</sup> pull			84/78	70/11	78/12	60/42
Durability % Failure						
25 cycles	90	95	0	0	0	0
100 cycles	98	98	0	0	0	40

[0048] Table 3 shows that waxes with melting points of 116°C or higher or an acid number of 40 or higher do not give adequate paint adhesion or durability (Comparative Examples 4-6).

#### Examples 8-9 and Comparative Examples 7-9

[0049] These examples and comparative examples show the effect of using oxidized polyethylene waxes with varying melting points on compositions containing MA-g-PP and an ATPEO in addition to the TPO, but no MA-g-rubber.

[0050] The TPO, MA-g-PP, ATPEO, and the antioxidant were the same as in Examples 1-4. The results are shown in Table 4. The melting points and other properties of the oxidized polyethylene waxes are shown in Table 2.

Table 4

	Comp. 7	Comp. 8	Comp. 9	Ex. 8	Ex. 9
TPO 1	100	100	100	100	100
MA-g-PP	10	10	10	10	10
Petr. E-1040	--	--	10	--	--
Petr. C-7500	--	--	--	10	--
Petr. C-8500	--	--	--	--	10
AC 307	10	--	--	--	--
AC 325	--	10	--	--	--
ATPEO	3	3	3	3	3
Antioxidant	0.2	0.2	0.2	0.2	0.2
Paint Adhesion					
% Failure	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	60/100	86/90	54/0	6/0	0/0
3 <sup>rd</sup> pull			70/26	48/0	36/0



Table 4 (continued)

	Comp. 7	Comp. 8	Comp. 9	Ex. 8	Ex. 9
Paint Adhesion					
5 <sup>th</sup> pull			70/54	72/0	60/12
Durability					
% Failure					
25 cycles	0	15	0	0	0
100 cycles	0	60	0	0	0

[0051] Table 4 shows that good adhesion and durability can be achieved if an oxidized polyethylene wax having a melting point below 116°C as well as an acid number less than 40 is used (Examples 8 and 9).

#### Examples 10-12 and Comparative Examples 10-11

[0052] These examples and comparative examples show the effect of using an oxidized polyethylene wax in a thermoplastic polyolefin composition containing two different heterophasic polyolefins, a metallocene rubber, MA-g-PP, and a functionalized polymer that reacts with the MA-grafted polymers, with and without a MA-g-rubber. The compositions also contain talc and conductive carbon black as fillers. The results are shown in Table 5.

[0053] In Table 5, heterophasic polyolefin 1 contained 85-86% polypropylene and 14-15% of an amorphous ethylene/propylene copolymer, of which 57% was ethylene and 43% was propylene and is commercially available from Montell USA Inc. Heterophasic polyolefin 2 contained 86.4% polypropylene, 9.45% of an amorphous ethylene/propylene copolymer that was soluble in xylene at room temperature, and 4.15 % of a semi-crystalline ethylene/propylene copolymer that was insoluble in xylene at room temperature and is commercially available from Montell USA Inc. The MA-g-PP and the MA-g-rubbers were described in Examples 1-4.

[0054] Engage™ 8150 ethylene/octene copolymer rubber contains 25% octene and is commercially available from DuPont-Dow Elastomers. HPVM 2203 hydroxy-terminated polybutene is commercially available from Shell Chemical Company.

[0055] The carbon black concentrate was Colonial™ 3300 carbon black, a concentrate of 50% N-100 black in low density polyethylene, commercially available from Colonial Rubber Company. The talc was Polar 9603 talc, commercially available from Polar Minerals. The antioxidant and the ATPEO were the same as in Examples 1-4.

Table 5

	Comp. 10	Comp. 11	Ex. 10	Ex. 11	Ex. 12
Het. Polyolefin 1	50	50	50	50	50
Het. Polyolefin 2	30	30	30	30	30
Engage 8150	20	20	20	20	20
MA-g-PP	10	10	10	10	10
MA-g-rubber 1	5	5	5	--	--
MA-g-rubber 2	5	5	5	--	--
Petr. C-3500	--	--	5	--	--
Petr. C-9500	--	--	--	5	10
Hydrox. polybutene	--	--	--	2	2
ATPEO	3	4	3	3	3
Talc	2	2	2	2	2
Carbon black conc.	4	4	4	4	4
Antioxidant	0.2	0.2	0.2	0.2	0.2

Table 5 (continued)

	Comp. 10	Comp. 11	Ex. 10	Ex. 11	Ex. 12
Paint Adhesion					
% Failure	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	100/100	100/100	46/8	40/0	32/0
3 <sup>rd</sup> pull			60/20	66/20	42/16
5 <sup>th</sup> pull			80/30	80/48	62/42
Durability % Failure					
25 cycles	100	100	0	0	0
100 cycles	100	100	0	18	12

[0056] Comparative Examples 10 and 11 without the waxes showed very poor paint adhesion and durability. Using the waxes with the MA-g-PP, with or without the MA-g-rubber, gave good adhesion and durability. Examples 11 and 12 show that a combination of an ATPEO and hydroxylated polybutene can be used as the functionalized polymer component of the composition.

#### Examples 13-17 and Comparative Examples 12-14

[0057] These examples and comparative examples show the effect of using an oxidized polyethylene wax in compositions containing two different thermoplastic polyolefins, MA-g-PP or MA-g-E/P, and a functionalized polymer that reacts with the MA-grafted polymers, with and without a MA-g-rubber. The results are shown in Table 6.

[0058] In Table 6, TPO 2 contained 68% of a ethylene/propylene copolymer having an ethylene content of 2.6%, 2% of a semi-crystalline ethylene/propylene copolymer that was insoluble in xylene at room temperature, and 30% of an amorphous ethylene/propylene copolymer that was soluble in xylene at room temperature.

[0059] TPO 1, the MA-g-PP, and MA-g-rubber 1 were described in Examples 1-4.

[0060] MA-g-E/P was ACX 597 maleic anhydride-grafted ethylene/propylene copolymer, commercially available from Allied Signal Inc. In Examples 16 and 17 and Comparative Examples 13 and 14, the MA-g-E/P was added in the form of a 10/3 or 10/4 adduct with the ATPEO, respectively. The ATPEO and the antioxidant were the same as in Examples 1-4.

Table 6

	Comp. 12	Comp. 13	Comp. 14	Ex. 13	Ex. 14	Ex. 15	Ex. 16	Ex. 17
TPO 1	--	100	100	--	100	100	100	100
TPO 2	100	--	--	100	--	--	--	--
MA-g-rubber 1	10	--	--	10	10	--	10	10
MA-g-PP	10	--	--	10	--	--	--	--
MA-g-E/P	--	--	--	--	10	10	--	--
10/3 Adduct	--	13	--	--	--	--	13	--
10/4 Adduct	--	--	14	--	--	--	--	14
Petr. C-9500	--	--	--	--	--	10	--	--
Petr. C-3500	--	--	--	5	5	--	5	5
ATPEO	3	--	--	3	3	3	--	--
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion								
% Failure	g/op	g/op	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	70/18	85/62	84/58	36/0	42/8	24/6	42/8	36/6

Table 6 (continued)

	Comp. 12	Comp. 13	Comp. 14	Ex. 13	Ex. 14	Ex. 15	Ex. 16	Ex. 17
Paint Adhesion								
3 <sup>rd</sup> pull	100/32	100/82	100/74	54/9	48/3 6	48/24	54/3 6	42/12
5 <sup>th</sup> pull		.. /100	.. /100	78/2 4	82/42	82/48	82/4 2	56/36
Durability % Failure								
25 cycles	70	100	100	0	10	16	32	35
100 cycles	80	100	100	22	35	30	38	42

[0061] Comparative Examples 12-14 without the waxes showed very poor paint adhesion and durability. Using the waxes with the MA-g-PP or MA-g- E/P, with or without the MA-g-rubber, gave good adhesion and durability.

#### Examples 18-22 and Comparative Example 15

[0062] These examples and the comparative example show the effect of using an oxidized polyethylene wax in a thermoplastic polyolefin composition containing MA-g-PP, a functionalized polymer that reacts with MA-grafted polymers, and talc as a filler, with and without a MA-g-rubber. The results are shown in Table 7.

[0063] In Table 7, the hydroxy-terminated polybutadiene was PolyBD R-45HT hydroxy-terminated polybutadiene, commercially available from Elf Atochem. TPO 1, the MA-g-rubbers and the MA-g-PP were described in Examples 1-4. The talc was Jetfill 700C talc, commercially available from Luzenac America. The hydroxy-terminated polybutene and the talc were described in Examples 10-12. The antioxidant and the ATPEO were the same as in the previous examples.

Table 7

	Comp. 15	Ex. 18	Ex. 19	Ex. 20	Ex. 21	Ex. 22
TPO 1	100	100	100	100	100	100
MA-g-rubber 1	5	5	5	5	--	--
MA-g-PP	10	10	10	10	10	10
MA-g-rubber 2	5	--	--	--	--	--
Petr. C-9500	--	--	--	--	10	10
Petr. C-3500	--	5	10	10	--	--
Talc	10	10	10	10	20	10
Hydr. polybutadiene	--	--	--	3	2	--
Hydr. polybutene	--	--	--	--	--	2
ATPEO	3	3	3	--	3	3
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion						
% Failure	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	98/48	3/0	0/0	0/0	1/0	0/3
3 <sup>rd</sup> pull	.. /100	26/0	0/6	9/0	1/0	24/16
5 <sup>th</sup> pull		54/3	6/14	16/6	1/0	42/42
Durability % Failure						
25 cycles	95	0	0	0	0	0
100 cycles	95	0	0	0	0	0

[0064] Comparative Example 15 without a wax showed very poor paint adhesion and durability. Using a wax with the MA-g-PP, with or without a MA-g-rubber, gave good adhesion and durability (Examples 18-22). Examples 21 and 22 showed that a combination of an ATPEO and hydroxy-terminated polybutene or hydroxy-terminated polybutadiene can be used as the functionalized polymer component of the composition.

#### Examples 23-29 and Comparative Example 16

[0065] These examples and the comparative example show the effect of using an oxidized polyethylene wax in a thermoplastic polyolefin composition that contains MA-g-PP, an ATPEO or a hydroxy-terminated ethylene/ethylene oxide copolymer, with or without MA-g-rubber. Some of the compositions contain talc as a filler and all contain either conductive or non-conductive carbon black. The results are shown in Table 8.

[0066] In Table 8, the carbon black was the same as in Examples 10-12. Printex™ XE-2 conductive carbon black is commercially available from Degussa Corporation. Vulcan™ XC-72 and Vulcan™ PA-90 conductive carbon blacks are commercially available from Cabot Corporation. In Examples 27 and 29, Printex™ XE-2 conductive carbon black was added as a 23% dispersion in pelletized TPO 1 containing 0.2 pph B 225 antioxidant. The amount of carbon black in the dispersion is enclosed in parentheses. In Example 28, Vulcan™ XC-72 conductive carbon black was added as a 33% dispersion in pelletized TPO 1 containing 0.2 pph B 225 antioxidant. The amount of carbon black in the dispersion is enclosed in parentheses. (TPO 1) in Examples 27-29 indicates the amount of TPO 1 used as the matrix for the dispersions of conductive carbon blacks.

[0067] Hydroxy-terminated EEO 1 was Unithox™ 480 hydroxy-terminated ethylene/ethylene oxide copolymer having a molecular weight  $M_n$  of 2250 and a hydroxyl number of 22, and is commercially available from Petrolite Corporation. TPO 1, the MA-g-rubbers, and the MA-g-PP were described in Examples 1-4. The antioxidant and the ATPEO were the same as in the previous examples. The talc was the same as in Examples 18-22.

Table 8

	Comp. 16	Ex. 23	Ex. 24	Ex. 25	Ex. 26	Ex. 27	Ex. 28	Ex. 29
TPO 1	100	100	100	100	100	74	72	74
(TPO 1)	--	--	--	--	--	(26)	(28)	(26)
MA-g-rubber 1	5	10	10	10	10	10	12	10
MA-g-PP	10	10	10	10	10	10	12	10
MA-g-rubber 2	5	--	--	--	--	--	--	--
Petr. C-3500	--	5	5	10	10	10	--	10
Talc	--	10	10	--	--	--	--	20
Carbon black	--	2	--	--	--	--	--	--
Printex XE-2	2	--	2	--	--	--	--	--
Vulcan XC-72	--	--	--	10	--	--	--	--
Vulcan PA-90	--	--	--	--	10	--	--	--
Dispersed XE-2	--	--	--	--	--	(6)	--	(6)
Dispersed XC-72	--	--	--	--	--	--	(14)	--
ATPEO	3	3	3	3	-	3	3.6	3
Hydrox. E/EO 1	--	--	--	--	3	--	--	--
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2	0.24	0.2
Paint Adhesion								
% Failure	g/op	g/op	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	100/48	0/0	0/0	0/0	18/0	8/6	18/0	0/0
3 <sup>rd</sup> pull	./100	9/0	12/6	0/0	24/0	50/12	48/0	0/6
5 <sup>th</sup> pull		20/6	23/32	6/2	42/4	74/30	76/18	15/18

Table 8 (continued)

	Comp. 16	Ex. 23	Ex. 24	Ex. 25	Ex. 26	Ex. 27	Ex. 28	Ex. 29
Durability % Failure								
25 cycles	40	0	0	0	0	2	5	0
100 cycles	50	0	30	0	10	20	40	0

[0068] Comparative Example 16 without the wax showed very poor paint adhesion and durability. Using the wax with the MA-g-PP, the functionalized polymer, and a filler, with or without the MA-grafted rubber, gave good adhesion and durability. Example 26 showed that a hydroxy-terminated ethylene/ethylene oxide copolymer can be used as the functionalized polymer without the use of an ATPEO.

#### Examples 30-37

[0069] Examples 30-34 show that good paint adhesion and durability can be obtained from compositions containing a TPO that includes two heterophasic polyolefins and an ethylene/octene copolymer rubber, as well as MA-g-PP, MA-g-rubber, an oxidized polyethylene wax, either an ATPEO or a hydroxy-terminated E/EO copolymer as the functionalized polymer, and, optionally, conductive carbon black as a filler. The results are shown in Table 9.

[0070] Good paint adhesion and durability are also obtained from compositions containing TPO 1, MA-g-PP, MA-g-rubber, an oxidized polyethylene wax, a hydroxy-terminated polyethylene or hydroxy-terminated E/EO copolymer as the functionalized polymer, and, optionally, talc as a filler (Examples 35-37). The results are given in Table 9.

[0071] In Examples 32-34, the Vulcan™ XC-72 conductive carbon black was added as a 33% dispersion in pelletized TPO 1 containing 0.2 pph B 225 antioxidant. The amount of carbon black in the dispersion is enclosed in parentheses. (TPO 1) in Examples 32-34 indicates the amount of TPO 1 used as the matrix for the dispersions of conductive carbon black.

[0072] Heterophasic polyolefins 1 and 2 were described in Examples 10-12. TPO 1, MA-g-PP, and MA-g-rubber 1 were described in Examples 1-4. The conductive carbon blacks were described in Examples 23-29. The antioxidant and the ATPEO were the same as in the previous examples. The talc was the same as in Examples 18-22.

[0073] Hydroxy-terminated E/EO 1 was described in Examples 23-29. Hydroxy-terminated E/EO 2 was Unithox™ 580 hydroxy-terminated ethylene/ethylene oxide copolymer having a molecular weight  $M_n$  of 2400 and a hydroxyl number of 18, commercially available from Petrolite Corporation. The hydroxy-terminated polyethylene was Unilin™ 425 hydroxy-terminated polyethylene, commercially available from Petrolite Corporation.

Table 9

	Ex. 30	Ex. 31	Ex. 32	Ex. 33	Ex. 34	Ex. 35	Ex. 36	Ex. 37
TPO 1	--	--	--	--	--	100	100	100
(TPO 1)	--	--	(23.3)	(23.3)	(23.3)	--	--	--
Het. Polyolefin 1	50	50	38.3	38.3	38.3	--	--	--
Het. Polyolefin 2	30	30	23.3	23.3	23.3	--	--	--
Engage 8150	20	20	15.1	15.1	15.1	--	--	--
MA-g-PP	15	15	11.5	17.25	17.25	10	10	10
MA-g-rubber 1	15	15	11.5	17.25	17.25	5	10	5
Petr. C-3500	10	10	11.5	11.5	11.5	10	10	10
Vulcan XC-72	--	10	--	--	--	--	--	--
Dispersed XC-72	--	--	(10)	(10)	(10)	--	--	--
Hydrox. E/EO 1	--	--	--	--	4	--	--	--
Hydrox. E/EO 2	--	--	--	--	--	3	--	3
Hydrox. PE	--	--	--	--	--	--	3	--
Talc	--	--	--	--	--	--	--	20

Table 9 (continued)

	Ex. 30	Ex. 31	Ex. 32	Ex. 33	Ex. 34	Ex. 35	Ex. 36	Ex. 37
ATPEO	4	4	3	4	--	--	--	--
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion								
% Failure	g/op	g/op	g/op	g/op	g/op	g/op	g/op	G/op
1 <sup>st</sup> pull	22/0	22/0	0/0	0/0	0/0	0/0	18/0	0/3
3 <sup>rd</sup> pull	38/0	50/12	0/0	0/0	0/0	45/8	36/6	0/18
5 <sup>th</sup> pull	64/12	74/28	0/0	0/0	0/0	78/40	42/18	10/38
Durability % Failure								
25 cycles	0	0	0	0	0	0	0	0
100 cycles	10	0	0	0	2	0	0	0

Examples 38-40 and Comparative Examples 17-18

[0074] These examples and comparative examples show the effect of using an oxidized polyethylene wax in a thermoplastic polyolefin composition containing MA-g-PP and a functionalized polymer that reacts with the maleic anhydride groups of the grafted polymers, with and without MA-g-rubber. The results are given in Table 10.

[0075] In Table 10, TPO 3 was a blend of 40 parts of a crystalline propylene homopolymer having a melt flow rate of ~12 g/10 min and xylene solubles at room temperature of 4%, commercially available from Montell USA Inc.; 60 parts of an olefin polymer composition containing 68% by weight of an ethylene/1-butene copolymer, of which 82% was ethylene, having xylene solubles at room temperature of 35%, dispersed in 32% of a propylene homopolymer matrix having xylene solubles at room temperature of 2.3%; and 0.2 parts of Irganox™ B 225 antioxidant per hundred parts of polymer.

[0076] MA-g-rubber 1 and the MA-g-PP were described in Examples 1-4. Hydroxy-terminated E/EO 2 was described in Examples 30-37. The antioxidant and the ATPEO were the same as in the previous examples.

Table 10

	Comp. 17	Comp. 18	Ex. 38	Ex. 39	Ex. 40
TPO 3	100	100	100	100	100
MA-g-rubber I	--	--	10	15	15
MA-g-PP	10	20	10	15	15
Petr. C-3500	--	--	10	10	10
ATPEO	3	4	3	4	--
Hydrox. E/EO 2	--	--	--	--	4
Antioxidant	0.2	0.2	0.2	0.2	0.2
Paint Adhesion					
% Failure	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	72/12	85/0	32/6	12/0	18/0
3 <sup>rd</sup> pull	82/32	100/24	52/24	35/12	24/6
5 <sup>th</sup> pull	100/56		68/42	50/24	42/24
Durability					
% Failure					
25 cycles	90	100	0	12	18
100 cycles	95	100	24	32	36

[0077] Comparative examples 17 and 18 without a wax showed very poor paint adhesion and durability. Using a wax with the MA-g-PP and the MA-g-rubber gave good adhesion and durability. Example 40 shows that a hydroxy-terminated ethylene/ethylene oxide copolymer can be used as the functionalized polymer component of the composition without an ATPEO.

#### Examples 41-45 and Comparative Examples 19-20

[0078] These examples and comparative examples show the effect of using an oxidized polyethylene wax in two different thermoplastic polyolefins containing a MA-g-PP, an ATPEO or a hydroxy-terminated ethylene/ethylene oxide copolymer as the functionalized polymer, and, optionally, a MA-g-rubber. One of the compositions also contains talc as a filler. The results are shown in Table 11.

[0079] In Table 11, the TPO was either (1) a blend of a propylene homopolymer having a melt flow rate of ~5 dg/min and an isotactic index greater than 90, commercially available from Montell USA Inc., and Dutral™ 4038 ethylene/propylene/ethylidenenorbornene terpolymer rubber containing 4% ethylidenenorbornene, commercially available from Enichem America Inc., or (2) heterophasic polyolefin 3, which contained (a) 35% of a propylene homopolymer having an isotactic index, defined as the xylene insoluble fraction, of 97.5, (b) 6.9% of a semi-crystalline ethylene-propylene copolymer that was xylene insoluble at room temperature, and (c) 58.1% of an amorphous ethylene-propylene copolymer rubber that was xylene soluble at room temperature.

[0080] The MA-g-PP and MA-g-rubber 1 were described in Examples 1-4. Hydroxy-terminated E/EO 2 was described in Examples 30-37. The antioxidant and the ATPEO were the same as in the previous examples. The talc was the same as in Examples 18-22.

Table 11

	Comp. 19	Comp. 20	Ex. 41	Ex. 42	Ex. 43	Ex. 44	Ex. 45
Polypropylene	70	--	70	--	--	--	70
Dutral 4038	30	--	30	--	--	--	30
Het. polyolefin 3	--	100	--	100	100	100	--
MA-g-rubber 1	--	--	10	10	10	10	10
MA-g-PP	10	10	10	10	10	10	10
Petr. C-3500	--	--	10	5	10	10	10
Talc	--	--	--	--	--	--	10
ATPEO	3	3	3	3	3	--	--
Hydrox. E/EO 2	--	--	--	--	--	3	3
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion							
% Failure	g/op	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	78/24	46/12	26/6	12/0	6/0	12/3	0/0
3 <sup>rd</sup> pull	100/32	68/28	38/6	22/6	18/0	16/6	12/0
5 <sup>th</sup> pull	-/68	100/48	56/18	36/24	24/12	18/12	36/12
Durability % Failure							
25 cycles	42	24	0	8	0	0	0
100 cycles	76	68	15	24	8	6	0

[0081] Comparative Examples 19 and 20 without the wax showed very poor paint adhesion and durability. Using a wax with the MA-g-PP and the MA-g-rubber gave good adhesion and durability. Examples 44 and 45 showed that a hydroxy-terminated ethylene/ethylene oxide copolymer can be used as the functionalized polymer instead of an ATPEO.

## Examples 46-51

[0082] These examples show the effect of using an oxidized polyethylene wax in a thermoplastic polyolefin composition containing MA-g-PP, maleic anhydride-grafted polyethylene, and a functionalized polymer that reacts with the maleic anhydride groups of the grafted polymers, with and without MA-g-rubber. The results are given in Table 12.

[0083] In Table 12, Ceramer™ 67 maleic anhydride-grafted polyethylene had a molecular weight  $M_n$  of 655, a maleic anhydride content of 3.8%, and a melting point of 97°C, and is commercially available from Petrolite Corporation. Ceramer™ 1608 maleic anhydride-grafted polyethylene had a molecular weight  $M_n$  of 700, a maleic anhydride content of 12.7% by weight, and a melting point of 121°C, and is commercially available from Petrolite Corporation.

[0084] TPO 1, MA-g-rubber 1, and MA-g-PP were described in Examples 1-4. Hydroxy-terminated E/EO 1 was described in Examples 23-29. TPO 1 was in pelletized form and contained 0.2 pph Irganox™ B-225 antioxidant. The antioxidant and the ATPEO were the same as in previous examples.

Table 12

	Ex. 46	Ex. 47	Ex. 48	Ex. 49	Ex. 50	Ex. 51
TPO 1	100	100	100	100	100	100
MA-g-rubber 1	10	10	..	..	10	..
MA-g-PP	10	10	10	10	10	10
Petr. C-3500	10	10	10	10	10	10
Ceramer 67	10	..	10	..	10	..
Ceramer 1608	..	10	..	10	..	10
ATPEO	3	3	3	3	..	..
Hydr. E/EO 1	..	..	..	..	3	3
Antioxidant	0.2	0.2	0.2	0.2	0.2	0.2
Paint Adhesion						
% Failure	g/op	g/op	g/op	g/op	g/op	g/op
1 <sup>st</sup> pull	0/0	0/0	0/0	0/0	0/0	0/0
3 <sup>rd</sup> pull	12/0	6/0	8/0	12/0	0/0	6/0
5 <sup>th</sup> pull	18/0	19/0	12/8	12/0	12/0	24/0
Durability % Failure						
25 cycles	0	0	0	0	0	0
100 cycles	0	0	4	8	0	0

## Claims

1. A composition comprising, by weight:

- (1) 100 parts of a thermoplastic polyolefin comprising an olefin polymer having an isotactic index of at least 80 and an olefin polymer rubber, the thermoplastic polyolefin having a rubber content of at least 20%;
- (2) 5 to 20 parts per hundred parts of the thermoplastic polyolefin of a propylene homopolymer or propylene copolymer with ethylene or a  $C_4$ - $C_8$   $\alpha$ -olefin having an ethylene or  $\alpha$ -olefin content of 0.5% to 20%, grafted with an anhydride of an aliphatic  $\alpha,\beta$ -unsaturated dicarboxylic acid and having an anhydride content of 2% to 5%;
- (3) 3 to 20 parts per hundred parts of the thermoplastic polyolefin of an oxidized polyethylene wax having a melting point of less than 116°C and an acid number of less than 40;
- (4) a functionalized polymer that is reactive with the anhydride groups of the grafted polymers, selected from the group consisting of:

- (a) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated polyalkylene



glycol;

(b) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyolefin,

(c) 2 to 6 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polybutadiene;

(d) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated olefin/alkylene oxide copolymer;

(e) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a hydroxy-terminated polyalkylene oxide,

(f) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of a methoxy-terminated polyalkylene oxide,

(g) 2 to 8 parts per hundred parts of the thermoplastic polyolefin of an amine-terminated olefin/alkylene oxide copolymer, and

(h) mixtures thereof;

(5) optionally, 5 to 30 parts per hundred parts of the thermoplastic polyolefin of a polyolefin rubber grafted with an anhydride of an aliphatic  $\alpha$ ,  $\beta$ -unsaturated dicarboxylic acid, having an anhydride content of at least 0.3% but less than 3% and comprising a polymer of ethylene and a  $C_3$ - $C_8$   $\alpha$ -olefin, optionally containing 0.5% to 10% of a diene, which contains 30% to 70% ethylene; and

(6) optionally 5 to 20 parts per hundred parts of the thermoplastic polyolefin of an ethylene polymer grafted with an anhydride of an aliphatic  $\alpha$ ,  $\beta$ -unsaturated dicarboxylic acid, having an anhydride content of 1% to 16% and a molecular weight  $M_n$  of 500 to 5000, provided that at least 5 parts of anhydride-grafted polypropylene or propylene copolymer and 3 parts of oxidized polyethylene wax per hundred parts of the thermoplastic polyolefin are also present.

2. The composition of claim 1 wherein the thermoplastic polyolefin is a composition comprising, by weight:

(a) 10% to 60% of a propylene homopolymer having an isotactic index greater than 90, or a crystalline propylene copolymer with ethylene and/or a  $C_{4-8}$   $\alpha$ -olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(b) 30% to 60% of an amorphous ethylene-propylene or ethylene-butene copolymer, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and having an ethylene content of 30% to 70%;

(c) 2% to 20% of a semi-crystalline ethylene-propylene or ethylene-butene copolymer that is xylene insoluble at room temperature and having an ethylene content of greater than 75% but less than 92%; and

(d) 5% to 20% of an ethylene polymer having a density of 0.91 to 0.96 g/cm<sup>3</sup> and a melt index of 0.1 to 100 g/10 min.

3. The composition of claim 1 wherein the thermoplastic polyolefin is a composition comprising, by weight:

(a) 20% to 70% of a crystalline propylene homopolymer having an isotactic index greater than 90, or a crystalline propylene copolymer with ethylene and/or a  $C_{4-8}$   $\alpha$ -olefin having a propylene content greater than 85% and an isotactic index of greater than 85;

(b) 20% to 75% of an amorphous copolymer of ethylene selected from the group consisting of (i) ethylene/propylene, (ii) ethylene/butene-1, (iii) ethylene/octene-1 and (iv) mixtures thereof, optionally containing 1% to 10% of a diene, which is xylene soluble at room temperature and has an ethylene content of 30% to 70%; and

(c) 2% to 30% of a semi-crystalline copolymer of ethylene selected from the group consisting of (i) ethylene/propylene, (ii) ethylene/butene-1, (iii) ethylene/octene-1 and (iv) mixtures thereof, which is xylene insoluble at room temperature and has an ethylene content of greater than 90%.

4. The composition of claim 1 wherein the thermoplastic polyolefin is a composition comprising, by weight:

(a) at least one heterophasic polyolefin composition comprising:

(i) 90% to 55% of a propylene polymer material selected from the group consisting of a propylene homopolymer having an isotactic index greater than 90, and a crystalline copolymer of propylene and an  $\alpha$ -olefin of the formula  $CH_2=CHR$ , where R is H or  $C_2$ - $C_6$  alkyl, the  $\alpha$ -olefin being less than 10% of the copolymer, and

(ii) 10% to 45% of an elastomeric copolymer of propylene and an  $\alpha$ -olefin of the formula  $CH_2=CHR$ , where R is H or  $C_2$ - $C_6$  alkyl, the  $\alpha$ -olefin being 50% to 70% of the elastomeric copolymer, and 10% to 40% of the elastomeric copolymer being insoluble in xylene at ambient temperature, and

(b) 5 to 50 parts per hundred parts of (a) of an elastomeric copolymer of ethylene and a C<sub>3</sub>-C<sub>8</sub> α-olefin made with a metallocene catalyst.

5. The composition of claim 1 wherein the thermoplastic polyolefin is a composition comprising, by weight:

(a) 30% to 50% of a propylene homopolymer having an isotactic index greater than 90, and

(b) 70% to 50% of an olefin polymer composition comprising:

(i) 25% to 50% of a crystalline propylene homopolymer with a solubility in xylene at room temperature of less than or equal to 4%, or a crystalline copolymer of propylene with ethylene or a C<sub>4</sub>-C<sub>8</sub> α-olefin having an ethylene or α-olefin content of 0.5% to 3%, and a solubility in xylene at room temperature of less than or equal to 4%, and

(ii) 50% to 75% of an amorphous copolymer of ethylene and a C<sub>4</sub>-C<sub>8</sub> α-olefin, wherein the α-olefin content is 10% to 20%, and the copolymer is 10% to 40% soluble in xylene at room temperature.

6. The composition of claim 1 wherein the thermoplastic polyolefin is a composition comprising, by weight:

(a) 80% to 30% of a propylene homopolymer having an isotactic index greater than 90, and

(b) 20% to 70% of an elastomeric copolymer of ethylene and a C<sub>3</sub>-C<sub>8</sub> α-olefin, optionally containing 1% to 10% of a diene, and having an ethylene content of 30% to 70%.

7. The composition of claim 1 wherein the anhydride of the aliphatic α,β-unsaturated dicarboxylic acid is maleic anhydride.

8. The composition of claim 1 wherein the functionalized polymer is selected from the group consisting of (a) amine-terminated polyethylene oxide, (b) hydroxy-terminated polyethylene oxide and (c) a hydroxy-terminated ethylene/ethylene oxide copolymer.

## Patentansprüche

1. Zusammensetzung, enthaltend, bezogen auf das Gewicht:

(1) 100 Teile eines thermoplastischen Polyolefins, das ein Olefinpolymer mit einem isotaktischen Index von mindestens 80 und einen Olefinpolymerkautschuk enthält und einen Kautschukgehalt von mindestens 20% aufweist;

(2) pro hundert Teile des thermoplastischen Polyolefins 5 bis 20 Teile eines Propylenhomopolymers oder Propylenocopolymers mit Ethylen oder einem C<sub>4</sub>-C<sub>8</sub>-α-Olefin mit einem Ethylen- bzw. α-Olefin-Gehalt von 0,5 bis 20%, das mit einem Anhydrid einer aliphatischen α,β-ungesättigten Dicarbonsäure gepropft ist und einen Anhydridgehalt von 2 bis 5% aufweist;

(3) pro hundert Teile des thermoplastischen Polyolefins 3 bis 20 Teile eines oxidierten Polyethylenwachses mit einem Schmelzpunkt von weniger als 116°C und einer Säurezahl von weniger als 40;

(4) ein funktionalisiertes Polymer, das gegenüber den Anhydridgruppen der gepropften Polymere reaktiv ist und aus der Gruppe bestehend aus:

(a) pro hundert Teile des thermoplastischen Polyolefins 2 bis 6 Teilen eines aminterminierten Polyalkylenglykols;

(b) pro hundert Teile des thermoplastischen Polyolefins 2 bis 6 Teilen eines hydroxyterminierten Polyolefins;

(c) pro hundert Teile des thermoplastischen Polyolefins 2 bis 6 Teilen eines hydroxyterminierten Polybutadiens;

(d) pro hundert Teile des thermoplastischen Polyolefins 2 bis 8 Teilen eines hydroxyterminierten Olefin/Alkylenoxid-Copolymers;

(e) pro hundert Teile des thermoplastischen Polyolefins 2 bis 8 Teilen eines hydroxyterminierten Polyalkylenoxids;

(f) pro hundert Teile des thermoplastischen Polyolefins 2 bis 8 Teilen eines methoxyterminierten Polyalkylenoxids;

(g) pro hundert Teile des thermoplastischen Polyolefins 2 bis 8 Teilen eines aminterminierten Olefin/Alky-

lenoxid-Copolymers und  
(h) Gemischen davon

stammt;

(5) gegebenenfalls pro hundert Teile des thermoplastischen Polyolefins 5 bis 30 Teile eines Polyolefinkautschuks, der mit einem Anhydrid einer aliphatischen  $\alpha,\beta$ -ungesättigten Dicarbonsäure gepropft ist und einen Anhydridgehalt von mindestens 0,3%, aber weniger als 3% aufweist und ein Polymer von Ethylen und einem  $C_3$ - $C_8$ - $\alpha$ -Olefin, das gegebenenfalls 0,5 bis 10% eines Diens enthält und 30 bis 70% Ethylen enthält, enthält; und

(6) gegebenenfalls pro hundert Teile des thermoplastischen Polyolefins 5 bis 20 Teile eines mit einem Anhydrid einer aliphatischen  $\alpha,\beta$ -ungesättigten Dicarbonsäure gepropften Ethylenpolymers mit einem Anhydridgehalt von 1 bis 16% und einem Molekulargewicht  $M_n$  von - 500 bis 5000, mit der Maßgabe, daß pro hundert Teile des thermoplastischen Polyolefins außerdem mindestens 5 Teile anhydridgepropftes Polypropylen oder Propylencopolymer und 3 Teile oxidiertes Polyethylenwachs vorliegen.

2. Zusammensetzung nach Anspruch 1, bei der es sich bei dem thermoplastischen Polyolefin um eine Zusammensetzung handelt, die, bezogen auf das Gewicht:

(a) 10 bis 60% eines Propylenhomopolymers mit einem isotaktischen Index von mehr als 90 oder eines kristallinen Propylencopolymers mit Ethylen und/oder einem  $C_4$ - $C_8$ - $\alpha$ -Olefin mit einem Propylengehalt von mehr als 85% und einem isotaktischen Index von mehr als 85;

(b) 30 bis 60% eines amorphen Ethylen-Propylen- oder Ethylen-Buten-Copolymers, das gegebenenfalls 1 bis 10% eines Diens enthält, bei Raumtemperatur xylollöslich ist und einen Ethylengehalt von 30 bis 70% aufweist;

(c) 2 bis 20% eines teilkristallinen Ethylen-Propylen- oder Ethylen-Buten-Copolymers, das bei Raumtemperatur xylolunlöslich ist und einen Ethylengehalt von mehr als 75%, aber weniger als 92% aufweist; und

(d) 5 bis 20% eines Ethylenpolymers mit einer Dichte von 0,91 bis 0,96 g/cm<sup>3</sup> und einem Schmelzindex von 0,1 bis 100 g/10 min enthält.

3. Zusammensetzung nach Anspruch 1, bei der es sich bei dem thermoplastischen Polyolefin um eine Zusammensetzung handelt, die, bezogen auf das Gewicht:

(a) 20 bis 70% eines kristallinen Propylenhomopolymers mit einem isotaktischen Index von mehr als 90 oder eines kristallinen Propylencopolymers mit Ethylen und/oder einem  $C_4$ - $C_8$ - $\alpha$ -Olefin mit einem Propylengehalt von mehr als 85% und einem isotaktischen Index von mehr als 85;

(b) 20 bis 75% eines amorphen Copolymers von Ethylen aus der Gruppe bestehend aus (i) Ethylen/Propylen, (ii) Ethylen/Buten-1, (iii) Ethylen/Octen-1 und (iv) Gemischen davon, das gegebenenfalls 1 bis 10% eines Diens enthält, bei Raumtemperatur xylollöslich ist und einen Ethylengehalt von 30 bis 70% aufweist; und

(c) 2 bis 30% eines teilkristallinen Copolymers von Ethylen aus der Gruppe bestehend aus (i) Ethylen/Propylen, (ii) Ethylen/Buten-1, (iii) Ethylen/Octen-1 und (iv) Gemischen davon, das bei Raumtemperatur xylolunlöslich ist und einen Ethylengehalt von mehr als 90% aufweist;

enthält.

4. Zusammensetzung nach Anspruch 1, bei der es sich bei dem thermoplastischen Polyolefin um eine Zusammensetzung handelt, die, bezogen auf das Gewicht:

(a) mindestens eine heterophasige Polyolefinzusammensetzung, die

(i) 90 bis 55% eines Propylenpolymers aus der Gruppe bestehend aus einem Propylenhomopolymer mit einem isotaktischen Index von mehr als 90 und einem kristallinen Copolymer von Propylen und einem  $\alpha$ -Olefin der Formel  $CH_2=CHR$ , worin R für H oder  $C_2$ - $C_8$ -Alkyl steht, wobei das  $\alpha$ -Olefin weniger als 10% des Copolymers ausmacht, und

(ii) 10 bis 45% eines elastomeren Copolymers von Propylen und einem  $\alpha$ -Olefin der Formel  $CH_2=CHR$ , worin R für H oder  $C_2$ - $C_8$ -Alkyl steht, wobei das  $\alpha$ -Olefin 50 bis 70% des elastomeren Copolymers ausmacht und 10 bis 40% des elastomeren Copolymers bei Umgebungstemperatur xylolunlöslich sind,

enthält, und

(b) pro hundert Teile von (a) 5 bis 50 Teile eines mit einem Metallocenkatalysator hergestellten elastomeren

Copolymers von Ethylen und einem C<sub>3</sub>-C<sub>8</sub>- $\alpha$ -Olefin

enthält.

5. Zusammensetzung nach Anspruch 1, bei der es sich bei dem thermoplastischen Polyolefin um eine Zusammensetzung handelt, die, bezogen auf das Gewicht:

- (a) 30 bis 50% eines Propylenhomopolymers mit einem isotaktischen Index von mehr als 90 und  
(b) 70 bis 50% einer Olefinpolymerzusammensetzung, die

(i) 25 bis 50% eines kristallinen Propylenhomopolymers mit einer Xyloflöslichkeit bei Raumtemperatur kleiner gleich 4% oder eines kristallinen Copolymers von Propylen mit Ethylen oder einem C<sub>4</sub>-C<sub>8</sub>- $\alpha$ -Olefin mit einem Ethylen- bzw.  $\alpha$ -Olefin-Gehalt von 0,5 bis 3% und einer Xyloflöslichkeit bei Raumtemperatur kleiner gleich 4% und

(ii) 50 bis 75% eines amorphen Copolymers von Ethylen und einem C<sub>4</sub>-C<sub>8</sub>- $\alpha$ -Olefin mit einem  $\alpha$ -Olefin-Gehalt von 10 bis 20% und einer Xyloflöslichkeit bei Raumtemperatur von 10 bis 40%

enthält,

enthält.

6. Zusammensetzung nach Anspruch 1, bei der es sich bei dem thermoplastischen Polyolefin um eine Zusammensetzung handelt, die, bezogen auf das Gewicht:

- (a) 80 bis 30% eines Propylenhomopolymers mit einem isotaktischen Index von mehr als 90 und  
(b) 20 bis 70% eines elastomeren Copolymers von Ethylen und einem C<sub>3</sub>-C<sub>8</sub>- $\alpha$ -Olefin, das gegebenenfalls 1 bis 10% eines Diens enthält und einen Ethylengehalt von 30 bis 70% aufweist,

enthält.

7. Zusammensetzung nach Anspruch 1, bei der es sich bei dem Anhydrid der aliphatischen  $\alpha,\beta$ -ungesättigten Dicarbonsäure um Maleinsäureanhydrid handelt.

8. Zusammensetzung nach Anspruch 1, bei der das funktionalisierte Polymer aus der Gruppe bestehend aus (a) aminterminiertem Polyethylenoxid, (b) hydroxyterminiertem Polyethylenoxid und (c) einem hydroxyterminierten Ethylen/Ethylenoxid-Copolymer stammt.

Revendications

1. Composition comprenant, en poids :

1) 100 parties d'une polyoléfine thermoplastique comprenant un polymère d'oléfine ayant un indice isotactique d'au moins 80 et un caoutchouc en polymère d'oléfine, la polyoléfine thermoplastique ayant une teneur en caoutchouc d'au moins 20% ;

2) 5 à 20 parties pour cent parties de la polyoléfine thermoplastique d'un homopolymère de propylène ou d'un copolymère de propylène avec l'éthylène ou une  $\alpha$ -oléfine en C<sub>4</sub>-C<sub>8</sub> ayant une teneur en éthylène ou en  $\alpha$ -oléfine de 0,5% à 20%, greffé avec un anhydride d'un acide dicarboxylique aliphatique insaturé  $\alpha, \beta$  et ayant une teneur en anhydride de 2% à 5% ;

3) 3 à 20 parties pour cent parties de la polyoléfine thermoplastique d'une cire oxydée à base de polyéthylène ayant un point de fusion inférieur à 116° C et un indice d'acide inférieur à 40 ;

4) un polymère fonctionnalisé qui est réactif avec les groupes anhydride des polymères greffés, choisi parmi le groupe consistant en :

a) 2 à 6 parties pour cent parties de la polyoléfine thermoplastique d'un polyalkylène glycol à terminaison amine ;

b) 2 à 6 parties pour cent parties de la polyoléfine thermoplastique d'une polyoléfine à terminaison hydroxy ;

- c) 2 à 6 parties pour cent parties de la polyoléfine thermoplastique d'un polybutadiène à terminaison hydroxy ;  
 d) 2 à 8 parties pour cent parties de la polyoléfine thermoplastique d'un copolymère oléfine / oxyde d'alkylène à terminaison hydroxy ;  
 e) 2 à 8 parties pour cent parties de la polyoléfine thermoplastique d'un oxyde de polyakylène à terminaison hydroxy ;  
 f) 2 à 8 parties pour cent parties de la polyoléfine thermoplastique d'un oxyde de polyakylène à terminaison méthoxy ;  
 g) 2 à 8 parties pour cent parties de la polyoléfine thermoplastique d'un copolymère oléfine d'oxyde d'alkylène à terminaison amine et,  
 h) leurs mélanges.

5) Facultativement, 5 à 30 parties pour cent parties de la polyoléfine thermoplastique d'un caoutchouc de polyoléfine greffé avec un anhydride d'un acide dicarboxylique aliphatique  $\alpha$ ,  $\beta$  insaturé, ayant une teneur en anhydride d'au moins 0,3% mais inférieure à 3% et comprenant un polymère d'éthylène et d'une  $\alpha$  oléfine en  $C_3-C_8$ , contenant facultativement 0,5% à 10% d'un diène, qui contient 30% à 70% d'éthylène et

6) Facultativement, 5 à 20 parties pour cent parties de la polyoléfine thermoplastique d'un polymère d'éthylène greffé avec un anhydride d'un acide dicarboxylique aliphatique  $\alpha$ ,  $\beta$  insaturé, ayant une teneur en anhydride de 1 % à 16%, ayant une masse moléculaire en nombre ( $M_n$ ) de 500 à 5000, pourvu qu'au moins 5 parties du polypropylène ou du copolymère de propylène greffés avec un anhydride et 3 parties de la cire oxydée à base de polyéthylène pour cent parties de la polyoléfine soient également présentes.

2. Composition selon la revendication 1 dans laquelle la polyoléfine thermoplastique est une composition comprenant, en poids :

- a) 10% à 60% d'un homopolymère de propylène ayant un indice isotactique supérieur à 90, ou un copolymère cristallin de propylène avec l'éthylène et/ou une  $\alpha$ -oléfine en  $C_4-C_8$  ayant une teneur en propylène supérieure à 85% et un indice isotactique supérieur à 85 ;  
 b) 30% à 60% d'un copolymère amorphe éthylène-propylène ou éthylène-butène, contenant facultativement 1% à 10% d'un diène, qui est soluble dans le xylène à température ambiante et ayant une teneur en éthylène de 30% à 70%  
 c) 2% à 20% d'un copolymère semi-cristallin éthylène-propylène ou éthylène-butène qui est insoluble dans le xylène à température ambiante et ayant une teneur en éthylène de plus de 75% mais de moins de 92% et  
 d) 5% à 20 % d'un polymère d'éthylène ayant une densité de 0,91 à 0,96 g/cm<sup>3</sup> et un indice de fluidité de 0.1 à 100 g/10 min.

3. Composition selon la revendication 1 dans laquelle la polyoléfine thermoplastique est une composition comprenant, en poids :

- a) 20% à 70% d'un homopolymère cristallin de propylène ayant un indice isotactique supérieur à 90, ou un copolymère cristallin de propylène avec l'éthylène et/ou une  $\alpha$ -oléfine en  $C_4-C_8$  ayant une teneur en propylène supérieure à 85% et un indice isotactique supérieur à 85 ;  
 b) 20% à 75% d'un copolymère amorphe d'éthylène choisi parmi le groupe constituant en (i) éthylène / propylène, (ii) éthylène / butène-1, (iii) éthylène / octène-1 et (iv) de leurs mélanges, contenant facultativement 1% à 10 % d'un diène, qui est soluble dans le xylène à température ambiante et qui a une teneur en éthylène de 30% à 70%, et  
 c) 2% à 30% d'un copolymère d'éthylène semi cristallin choisi parmi le groupe constituant en (i) éthylène / propylène, (ii) éthylène / butène-1, (iii) éthylène / octène-1, et (iv) de leurs mélanges, qui est insoluble dans le xylène à température ambiante et à une teneur en éthylène de plus de 90%

4. La composition selon la revendication 1 dans laquelle la polyoléfine thermoplastique est une composition comprenant, en poids :

- a) au moins une composition de polyoléfine hétérophasique comprenant :

i) 90% à 95% d'une matière polymère de propylène choisi parmi le groupe constituant en un homopolymère de propylène, ayant un indice isotactique supérieur à 90 et un copolymère cristallin de propylène, et d'une  $\alpha$ -oléfine répondant à la formule  $CH_2=CHR$ , dans laquelle R représente un atome d'hydrogène

ou un groupe alkyle en C<sub>2</sub>-C<sub>8</sub>, l' $\alpha$ -oléfine représentant moins de 10% du copolymère, et  
 ii) 10% à 45% d'un copolymère élastomère de propylène et d'une  $\alpha$ -oléfine répondant à la formule  
 CH<sub>2</sub>=CHR, dans laquelle R représente un atome d'hydrogène ou un groupe alkyle en C<sub>2</sub>-C<sub>8</sub>, l' $\alpha$ -oléfine  
 représentant 50% à 70% du copolymère élastomère, et 10% à 40% du copolymère élastomère étant  
 insoluble dans le xylène à température ambiante, et

b) 5 à 50 parties pour cent parties de (a) d'un copolymère élastomère d'éthylène et d'une  $\alpha$ -oléfine en C<sub>3</sub>-C<sub>8</sub>  
 fabriqué avec un catalyseur métallocène.

5. Composition selon la revendication 1 dans laquelle la polyoléfine thermoplastique est une composition compre-  
 nant, en poids :

a) 30% à 50% d'un homopolymère de propylène ayant un indice isotactique supérieur à 90, et  
 b) 70% à 50 % d'une composition de polymère d'oléfine comprenant :

i) 25% à 50% d'un homopolymère cristallin de propylène avec une solubilité dans le xylène à température  
 ambiante inférieure ou égale à 4% ou d'un copolymère cristallin de propylène avec l'éthylène ou une  $\alpha$ -  
 oléfine en C<sub>4</sub>-C<sub>8</sub>, ayant une teneur en éthylène ou en  $\alpha$ -oléfine de 0,5% à 3% et une solubilité dans le  
 xylène à température ambiante inférieure ou égale à 4 % et,

ii) 50% à 75% d'un copolymère amorphe d'éthylène et d'une  $\alpha$ -oléfine en C<sub>4</sub>-C<sub>8</sub> dans lequel la teneur en  
 $\alpha$ -oléfine est de 10% à 20%, le copolymère étant, soluble de 10% à 40% dans le xylène à température  
 ambiante

6. Composition selon la revendication 1 dans laquelle la polyoléfine thermoplastique est une composition compre-  
 nant, en poids :

a) 80% à 30% d'un homopolymère de propylène ayant un indice isotactique supérieur à 90, et  
 b) 20% à 70% d'un copolymère élastomère d'éthylène et d'une  $\alpha$ -oléfine en C<sub>3</sub>-C<sub>8</sub>, contenant facultativement  
 1% à 10% d'un diène, et ayant une teneur en éthylène de 30% à 70 %.

7. Composition selon la revendication 1 dans laquelle l'anhydride de l'acide dicarboxylique  $\alpha$ ,  $\beta$  insaturé est l'anhy-  
 dride maléique.

8. Composition selon la revendication 1 dans laquelle le polymère fonctionnalisé est choisi parmi le groupe constituant  
 en a) oxyde de polyéthylène à terminaison amine, b) oxyde de polyéthylène à terminaison hydroxy et c) un copo-  
 lymère d'oxyde d'éthylène/ éthylène à terminaison hydroxy.